

# An evaluation of ozone exposure metrics for a seasonally drought-stressed ponderosa pine ecosystem

Jeanne A. Panek\*, Meredith R. Kurpius, Allen H. Goldstein

*Environmental Science, Policy and Management, University of California, 151 Hilgard Hall, Berkeley, CA 94720, USA*

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**“Capsule”:** *Use of the SUM0 metric (sum of all daytime ozone concentrations) was only appropriate early in the growing season for ponderosa pine but overestimated ozone uptake under drought conditions later in the season.*

## Abstract

Ozone stress has become an increasingly significant factor in cases of forest decline reported throughout the world. Current metrics to estimate ozone exposure for forest trees are derived from atmospheric concentrations and assume that the forest is physiologically active at all times of the growing season. This may be inaccurate in regions with a Mediterranean climate, such as California and the Pacific Northwest, where peak physiological activity occurs early in the season to take advantage of high soil moisture and does not correspond to peak ozone concentrations. It may also misrepresent ecosystems experiencing non-average climate conditions such as drought years. We compared direct measurements of ozone flux into a ponderosa pine canopy with a suite of the most common ozone exposure metrics to determine which best correlated with actual ozone uptake by the forest. Of the metrics we assessed, SUM0 (the sum of all daytime ozone concentrations > 0) best corresponded to ozone uptake by ponderosa pine, however the correlation was only strong at times when the stomata were unconstrained by site moisture conditions. In the early growing season (May and June), SUM0 was an adequate metric for forest ozone exposure. Later in the season, when stomatal conductance was limited by drought, SUM0 overestimated ozone uptake. A better metric for seasonally drought-stressed forests would be one that incorporates forest physiological activity, either through mechanistic modeling, by weighting ozone concentrations by stomatal conductance, or by weighting concentrations by site moisture conditions. © 2002 Elsevier Science Ltd. All rights reserved.

**Keywords:** Forest ozone injury; Ozone monitoring and assessment; Ozone exposure metrics: SUM0, SUM06, SUM08, W126, AOT40; Forest-atmosphere interactions

## 1. Introduction

Ozone is a common, phytotoxic air pollutant that has become progressively more prevalent in industrialized parts of the world and an increasingly significant factor in the decline of forest health. Ozone damage to forests has been reported worldwide, including forests in eastern and western Europe (Rennenberg et al., 1997; Matyssek et al., 1997), and throughout the United States (e.g. in California, Miller et al., 1998, in New England, Treshow, 1984, in the southeastern United States, Skelly et al., 1997). Cause and effect relationships are being sought between ozone exposure and

forest response. This requires a measure of biologically meaningful ozone exposure.

Ozone concentration is routinely monitored throughout the United States. Air pollution specialists have explored mathematical approaches for summarizing ambient air quality information into forms that can serve as a surrogate for dose (Lefohn, 1992). These ozone metrics are being used to describe forest exposure to ozone with the end of establishing cause and effect relationships in standing forests. Dose has been historically defined as concentration $\times$ time (O’Gara, 1922). Effective dose was further defined as the concentration adsorbed by vegetation (Runeckles, 1974) in contrast to ambient air concentration. Fowler and Cape (1982) recognized the role of stomatal conductance as a means for ozone to reach leaf internal surfaces and introduced pollutant adsorbed dose, defined in units of  $\text{g m}^{-2}$  ground or leaf area. Taylor et al. (1982) further added

\* Corresponding author. Tel.: +1-510-642-9732; fax: +1-510-643-5098.

E-mail address: jpanek@nature.berkeley.edu (J.A. Panek).

the term internal flux ( $\text{mg m}^{-2} \text{h}^{-1}$ ) as a measure of physiologically relevant ozone exposure.

Several indices have been developed to describe vegetation exposure to ozone. Some have evolved from the observed relationship that short-term high concentrations of ozone cause significant damage. Others characterize the deleterious effects of long-term moderate concentrations. The 7-h mean (09:00–15:59) was established by the United States EPA National Crop Loss Assessment Network (NCLAN) as the statistic that best represented the period of greatest plant vulnerability to ozone pollution and highest ozone concentrations (Heck et al., 1982; Lefohn, 1992). Later it was recognized that not all sites experience the maximum ozone concentration between those hours and the window was expanded to 12 h. In the late 1980s the EPA abandoned the use of a mean concentration statistic when it was recognized that means were inadequate in describing the ozone environment (Lee et al., 1988) and instead cumulative indices were introduced. Cumulative indices with various threshold levels ( $> 60$  ppb,  $> 80$  ppb) and indices designed to give greater weight to higher concentrations (e.g. W126, Lefohn and Benedict, 1982) were successful at capturing the variability in crop response to ozone (Lee et al., 1988) and have been adopted by the forest research community in an attempt to relate ozone exposure to observed injury. Throughout the United States, the indices that are routinely employed include SUM0 (the sum of all hourly ozone concentrations in a 14-h daytime period, expressed in ppb-h), SUM06 (the sum of daytime ozone concentration hours  $> 60$  ppb, ppb-h), SUM08 (the sum of daytime ozone concentration hours  $> 80$  ppb, ppb-h) and W126 (an index derived from sigmoidally weighting ozone concentrations, ppb-h). In Europe, the index AOT40 (the sum of all daytime ozone concentrations above 40 ppb) has become the standard monitoring metric. Although the role of stomatal conductance in influencing the effective dose of ozone is generally recognized (Hogsett et al., 1989; Runeckles, 1992), the physiological activity of the plant has not been explicitly incorporated into ozone exposure indices in either the United States or Europe. Stomata are the entry point for ozone into the leaf. Most damage occurs once ozone gets inside the leaf (Reich, 1987; Darvall, 1989; Runeckles and Chevone, 1992; Weber et al., 1993). Stomatal aperture in trees is influenced by environmental factors, including light, soil water availability and atmospheric humidity and is under tight biological control. In many parts of the United States and Europe, trees and crops are well-watered and therefore active most of the growing season. In these places ozone concentration and concentration-derived indices probably correlate well with ozone flux into plants. This, however, may be an invalid assumption in the forests of most of California and the Pacific Northwest. It may also be invalid in ecosystems experiencing non-average

climate conditions, like stressful drought years. The ozone monitoring community is debating the utility of standard metrics as general tools for the monitoring and assessment of forest health, recognizing that concentration indices may be poor metrics of ozone uptake (in Europe e.g. Emberson et al., 2000; Fuhrer, 2000; in North America, e.g. Legge et al., 1995; Musselman and Massman, 1999; Massman et al., 2000). With this paper, we hope to contribute to that debate and advance the understanding of the utility of common metrics. Until now, there have been very few datasets which could address this concern, especially in a seasonally drought-stressed ecosystem.

In the Mediterranean climate of California, ozone is a significant pollutant during the hot and dry summer months. Ponderosa pine are particularly sensitive to soil moisture deficits and respond to the protracted summer drought by progressively constraining stomatal conductance throughout the season (Running, 1976; Bassman, 1988; Goldstein et al., 2000; Panek and Goldstein, 2001). When stomata are constrained, ozone movement into the foliage is limited. Recent studies of ponderosa pine decline in California have used the standard ozone metrics and have taken advantage of existing gradients of ozone to explore the relationship between injury and exposure. Of the various measures of ozone exposure, SUM0 and W126 were the metrics most strongly related to injury (Miller et al., 1998; Arbaugh et al., 1998; Salardino and Carroll, 1998). Injury was determined using an ozone injury index (OII), which incorporates alterations to phenology and an estimate of visible chlorotic mottling. Miller et al. (1998) found that SUM0 was best correlated with chlorotic mottle and fascicle retention ( $R^2=0.57$ ,  $R^2=0.74$ , respectively) at sites across the Sierra Nevada and San Bernardino transect. Salardino and Carroll (1998) found the correlation between damage and SUM0 not to be as good for only 2 years of the Sierra Nevada study ( $R^2=0.59$  for chlorotic mottle,  $R^2=0.39$  for fascicle retention). Arbaugh et al. (1998) were able to increase the strength of the relationship between OII and SUM0 in the Sierra Nevada by including 4 years of data in their analysis ( $R^2=0.70$ ) and an extra “indicator variable” for number of trees with  $\geq 90\%$  injury ( $R^2=0.93$ ). This “indicator variable” allowed the southern, most-polluted sites to fit the trend, however its mechanistic relevance was unexplained in the paper. We believe we can explain why short-term relationships between SUM0 and OII were poor, while long-term relationships were better. It is unlikely that ozone concentration based indices will ever be powerful enough to develop short-term cause–effect relationships in natural ecosystems in California because of its Mediterranean climate. The timing of greatest forest physiological activity and therefore ozone uptake does not correspond to the period of highest ozone concentrations that profoundly influence the indices.

The disparity between ozone uptake and ozone concentration has been shown experimentally. Using the eddy covariance method to measure fluxes of ozone into a California ponderosa pine forest, we found that in 1997 periods of high ozone deposition and periods of high ozone concentration were decoupled from each other both seasonally and diurnally (Fig. 1, Bauer et al., 2000). Highest ozone deposition preceded highest ozone concentration by 2–3 h daily, and by about a month during the growing season. Ozone deposition velocity was highly correlated with stomatal conductance (Bauer et al., 2000). As a result, the period of maximum deposition occurred in early summer when the stomata were relatively unconstrained by drought stress, however the highest concentrations of ozone occurred in the late summer.

The purpose of this paper, was to test how well the metrics of ozone exposure currently used in forest monitoring in California and throughout North America compared with the flux of ozone into the forest. We compared direct eddy covariance measurements of ozone flux into a ponderosa pine canopy with the ozone concentration based metrics SUM0, SUM06, SUM08 and W126 over two growing seasons. We asked the following questions. What period of the growing season and under what conditions did the metrics perform best? How much error is incorporated into standard metrics relative to direct measures of ozone uptake?

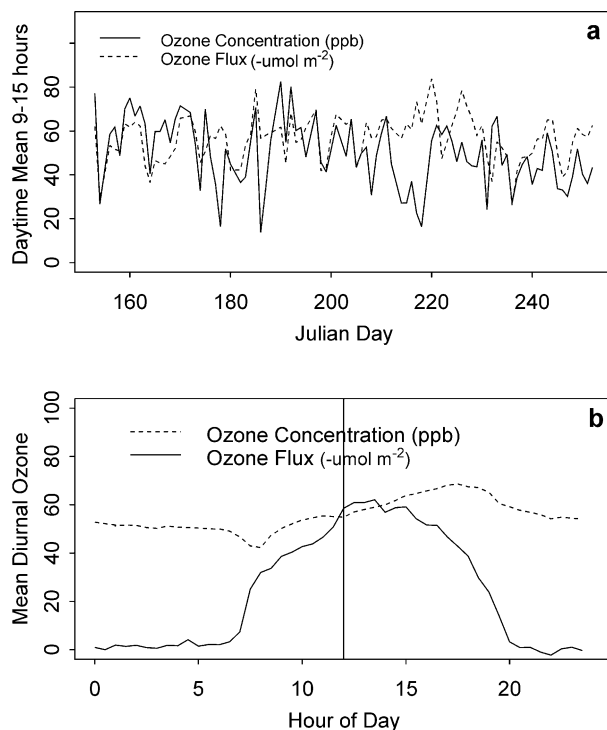


Fig. 1. 1997 data from the eddy flux tower site demonstrates that peak ozone flux is decoupled from peak ozone concentration, both seasonally (a) and diurnally (b). Redrawn from Bauer et al. (2000).

## 2. Materials and methods

### 2.1. Site description

Blodgett Forest Research Station is a 1200 ha mixed coniferous forest located in the middle elevation (1300–1500 m) of the central Sierra Nevada near Georgetown, California (38°53'42.9" N, 120°37'57.9" W), managed by the University of California. Ecosystem scale flux measurements were made in a ponderosa pine plantation on land owned by Sierra Pacific Industries, adjacent to Blodgett Forest (details in Goldstein et al., 2000). Typical wind patterns at the site included upslope flow during the day carrying polluted air masses from the Sacramento Valley, and cleaner downslope flow at night. The plantation was relatively flat, and contained a homogenous mixture of 5–7-year-old ponderosa pine with other trees and shrubs scattered throughout the ecosystem making up less than 30% of the biomass in 1997. In 1998 shrub cover grew to about 30% of the biomass. Ozone flux was measured 9 m above the ground and 5 m above the canopy using eddy covariance (Bauer et al., 2000; Goldstein et al., 2000), a technique that measures the flux of a scalar at a point centered on instruments placed at some height above the surface. Ozone flux,  $F_{O_3}$ , was calculated as:

$$F_{O_3} = \overline{w'O_3'}$$

where  $w$  is the vertical wind velocity and  $O_3$  the concentration of ozone at the measurement height. The prime indicates instantaneous deviation from the mean and the overbar indicates the time average of 30 min. Positive flux represents mass transfer from the surface into the atmosphere while negative flux represents mass transfer from the atmosphere into the surface; ozone flux is typically negative. The data acquisition system was separated into two main parts: (1) a slow response system that stored  $O_3$  concentration data averaged over 30 min intervals (measured by a Dasibi 1008) and (2) a fast response system that collected ozone, wind speed and wind direction data at 10 Hz which was used to calculate trace gas and energy fluxes by eddy covariance. Fast response measurements of ozone were made by chemiluminescence using Coumarin dye with an instrument custom built by NOAA, and wind speed and direction in three dimensions was measured by a sonic anemometer (Applied Technologies, Inc., Boulder, CO). The precision of the Dasibi ozone instrument is 1 ppb and yearly factory calibrations have confirmed its accuracy to within 1%.

Soil moisture was measured using time domain reflectometry (Campbell Scientific Inc.). In 1997, soil moisture was measured at 10 and 20 cm below the soil surface. In 1998, soil moisture was measured in a different location at 10, 30, and 50 cm below the soil surface.

Ozone concentration, flux and soil moisture were measured from 1 June to 9 September 1997 (days 152–252) and from 1 May to 31 October 1998 (days 121–304; Bauer et al., 2000; Goldstein et al., 2000).

## 2.2. Ozone metrics

We present 2 years of ozone data, one representing a dry growing season (1997) and one representing a wet growing season (1998). We calculated the ozone concentration metrics used in forest monitoring networks in California (Miller et al., 1998; Arbaugh et al., 1998; Salardino and Carroll, 1998). All metrics were constrained to the daylight hours between 06:00 and 19:59 because 14-h exposure indices were found to correlate better with OII than 7-h indices (09:00–15:59) in California (Salardino and Carroll, 1998, Miller internal reports). W126 is derived by sigmoidally weighting ozone mean concentrations with the following function:

$$w_i = \frac{1}{[1 + M \times \exp(-A \times c_i)]}$$

where  $M$  and  $A$  are positive arbitrary constants (4403 and 126 ppm<sup>-1</sup>, respectively, Lefohn et al., 1988),  $w_i$  is the weighting factor for concentration and  $c_i$  is the ozone concentration in ppb. Using the eddy covariance data, we summed the measured ozone flux over the 06:00–19:59 daylight period, SUMFLUX (umol m<sup>-2</sup> h<sup>-1</sup> × 14 h), to compare with ozone metrics over the same time period.

We then compared the ozone metrics against measured ozone flux values daily using least means regression. The 1998 growing season was particularly wet (rainfall from 1 February to 1 June was 176% above normal), while the 1997 season was dry (27% below normal). The metrics showed a marked change in the ability to predict flux after soil moisture had reached its minimum in both years. So, we divided the season into two periods of differing soil moisture, representing soil moisture well above a minimum and soil moisture near or at a minimum (Fig. 2). In 1997 soil moisture probes were installed two weeks after we started flux measurements. The soil moisture during that time was modeled using rainfall data and known soil drying curves.

## 3. Results

### 3.1. 1998 growing season

The 1998 season was the most informative because it included a broad range of moisture. Soil moisture is known to affect stomatal conductance at this site through its effects on internal plant water potential (Panek and Goldstein, 2001), and as stomata are the

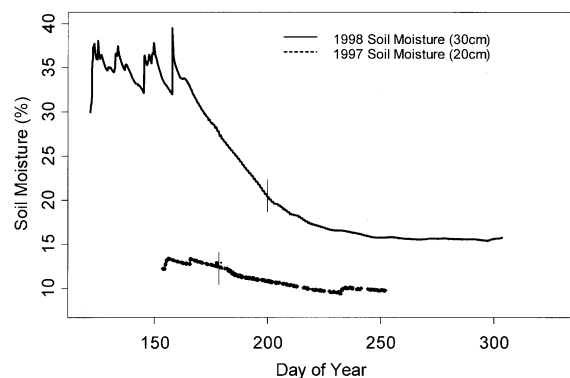


Fig. 2. Soil moisture compared for the 1997 and 1998 growing seasons. Vertical lines indicate where data were divided into “above minimum” and “near or at minimum”.

main sink for ozone (Bauer et al., 2000) the effect of soil moisture on the relationship between ozone metrics and ozone uptake was not a surprise. The best metric of ozone flux was SUM0 (Table 1, Fig. 3). The relationship was best early in the season when soil moisture values were greater than 20% ( $R^2=0.79$ ). Later in the season, at soil water values less than 20%, SUM0 was not well correlated with ozone flux (Figs. 3 and 5). Likewise, W126 and SUM06 were best correlated with ozone flux early in the season, although the relationships were weak ( $R^2=0.53$ , 0.46, respectively). The strength of the correlation decreased with decreasing soil moisture (Table 1, Figs. 3 and 5). SUM08 was not correlated with ozone flux at any time during the season (Table 1).

The relationship between all the metrics over the course of the growing season is shown in Fig. 5. The parallel trend of ozone flux and SUM0 at the beginning of the season was clear. Initially, ozone concentrations were low and values rarely exceeded 60 ppb. In the

Table 1

Results for linear regression of ozone concentration metrics against ozone flux into a ponderosa pine canopy in the 1998 and 1997 growing seasons

Metric	Soil moisture	Ozone flux 1998			Ozone flux 1997		
		$R^2$	$P$ -value	$N$	$R^2$	$P$ -value	$N$
SUM0	Entire season	0.36	<0.0001	172	0.38	<0.0001	96
	> minimum	0.79	<0.0001	72	0.78	<0.0001	22
	at minimum	0.19	=0.0007	58	0.42	<0.0001	74
W126	Entire season	0.25	<0.0001	172	0.17	<0.0001	96
	> minimum	0.53	<0.0001	72	0.20	=0.0377	22
	at minimum	0.12	=0.0073	58	0.24	<0.0001	74
SUM06	Entire season	0.23	<0.0001	172	0.16	=0.0001	96
	> minimum	0.46	<0.0001	72	0.15	=0.0765	22
	at minimum	0.09	=0.0244	57	0.23	<0.0001	74
SUM08	Entire season	0.01	=0.1229	172	0.05	=0.028	96
	> minimum	0.05	=0.0693	72	0.04	=0.353	22
	at minimum	0.07	=0.066	58	0.08	=0.016	74

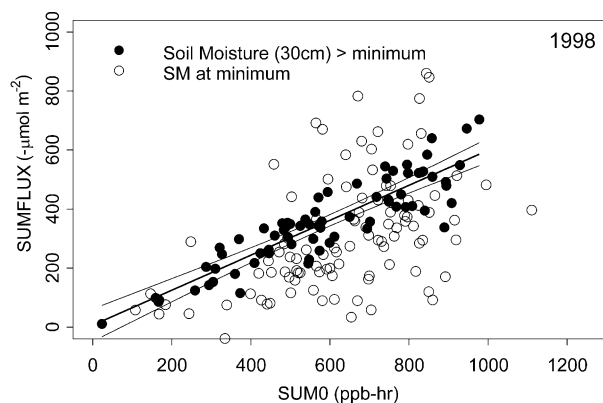


Fig. 3. The relationship between ozone flux into a ponderosa pine canopy (SUMFLUX) and the ozone exposure metric SUM0 for consecutive periods of differing soil moisture over the 1998 growing season. The regression line and the 95% confidence interval are shown.

mid- and late-seasons, ozone concentrations were frequently above 60 ppb and often above 80 ppb, however by day 205 ozone fluxes fell in response to lower soil moisture effects on stomata (Bauer et al., 2000). This decoupling between ozone flux and ozone concentration later in the season led to the poor performance of the metrics at that time.

### 3.2. 1997 growing season

The 1997 growing season was very dry, and was our first season of eddy flux measurements. Measurements started later in 1997 than in 1998, when the soil moisture had already dropped below 20%. Despite the low soil moistures, SUM0 predicted ozone flux well until soil moisture neared a minimum of 9% (Table 1, Figs. 4 and 6). At no time during the 1997 growing season were W126, SUM06 or SUM08 good predictors of the ozone flux (Table 1).

## 4. Discussion

The underlying assumption of this paper is that ozone must enter the leaf to cause damage and therefore flux into the canopy is the best standard of ozone exposure against which to compare exposure indices. This is supported by the general understanding that most, albeit not all (Bennett et al., 1973), ozone damage occurs within the leaf in the area surrounding the substomatal cavity (Runeckles, 1992; Runeckles and Chevone, 1992). Here, ozone forms highly toxic free radicals in the apoplast, within cell walls, and within plant cells that can damage most cell components (Runeckles and Chevone, 1992; Bytnerowicz, 1996). Thus, ozone flux is the best standard against which to compare the performance of concentration metrics.

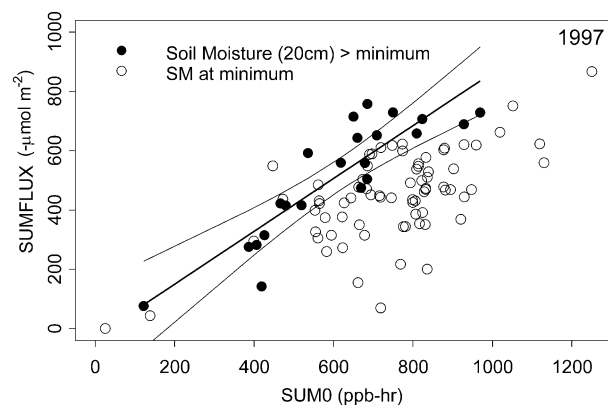


Fig. 4. The relationship between ozone flux into a ponderosa pine canopy (SUMFLUX) and the ozone exposure metric SUM0 for consecutive periods of differing soil moisture over the 1997 growing season. The regression line and the 95% confidence interval are shown.

SUM0 was the metric that was best correlated to ozone flux. The good correlation was limited to a period in the early growing season when stomata were less constrained by moisture. In 1998, a wet El Niño year, the relationship was strong through day 200 (19 July). In 1997, however, a drier year more typical of the Mediterranean climate of California, the relationship held only through day 175 (24 June). In both years the good relationship between SUM0 and flux held until soil moisture neared its minimum even though the minima in the two years were different. W126 was not as good at predicting ozone flux as SUM0. In no case was SUM06 or SUM08 a good metric for ozone flux. In general, in a typical year the ozone indices probably overestimate the ozone uptake by a forest canopy over the course of the growing season. In the early season, the indices are fairly representative since most of the ozone in the air is deposited into the foliage, but later in the season indices are based on ozone concentrations that are high but are not taken up by the canopy.

The strength of the correlation between SUM0 and ozone flux early in the season may in part explain the relationship between SUM0 and observed damage along an ozone injury gradient in California (Arbaugh et al., 1998). Of all the metrics, SUM0 best represents ozone uptake by the canopy, but only when stomata are not limited by moisture availability. In a wet year, this may be half the growing season. This may also explain why the metrics did not perform well in individual years (Miller internal reports) and in the short-term (Salardino and Carroll, 1998), but performed better when 4 years of data — including wet years as well as dry years — were included in the analysis (Arbaugh et al., 1998). The “indicator variable” used by Arbaugh et al. (1998) to fit the most-polluted southern site to the general trend may be a factor related to stomatal conductance. Preliminary conductance measurements at that site show that stomata were less constrained under drought stress than at

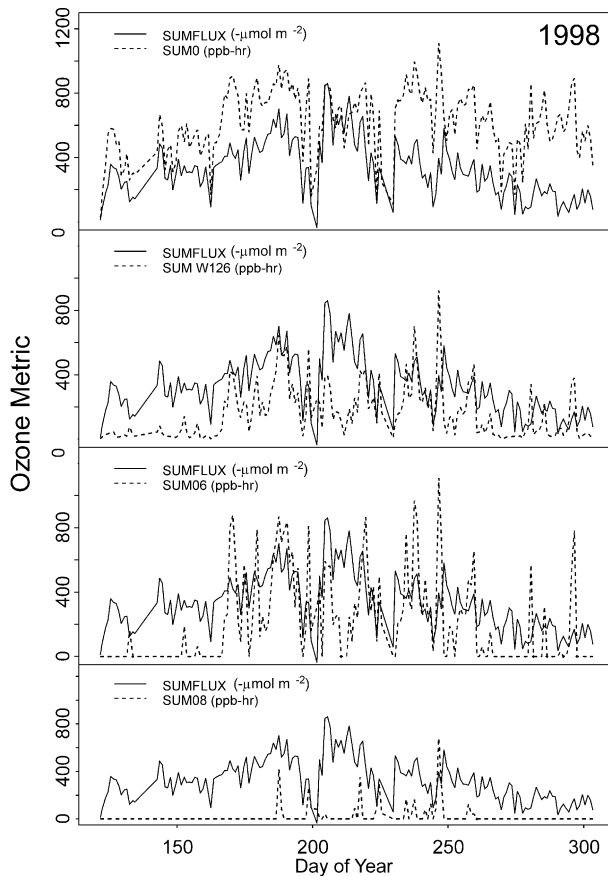


Fig. 5. A comparison of ozone metrics and ozone flux measured over the 1998 growing season.

the other sites and ozone fluxes into trees there were higher (Panek, personal observation).

Ozone must get into the plant before it can cause significant damage, however ozone flux still may not be highly correlated with ozone damage. Damage is the result of the plant's inability to effectively deal with the ozone stress. Trees have a host of mechanisms with which to cope with ozone injury — antioxidants, carbon stores for repair, allocation, etc. Musselman and Massman (1999) discuss this in terms of “effective flux” and “cumulative effective loading”. Furthermore, some individuals within a species are genetically more predisposed to ozone sensitivity than others. However, it will be much more difficult to understand how trees respond to the ozone environment in which they grow, unless the physiologically relevant ozone exposure to which they are subject is quantified. This is necessary to determine relationships between injury, protective responses, changes in metabolism and in allocation, and to determine whether pulses of high ambient air concentrations are getting into the leaves, or whether low concentrations during periods of peak physiological activity are more important. Understanding these relationships is vital in designing effective mitigation schemes.

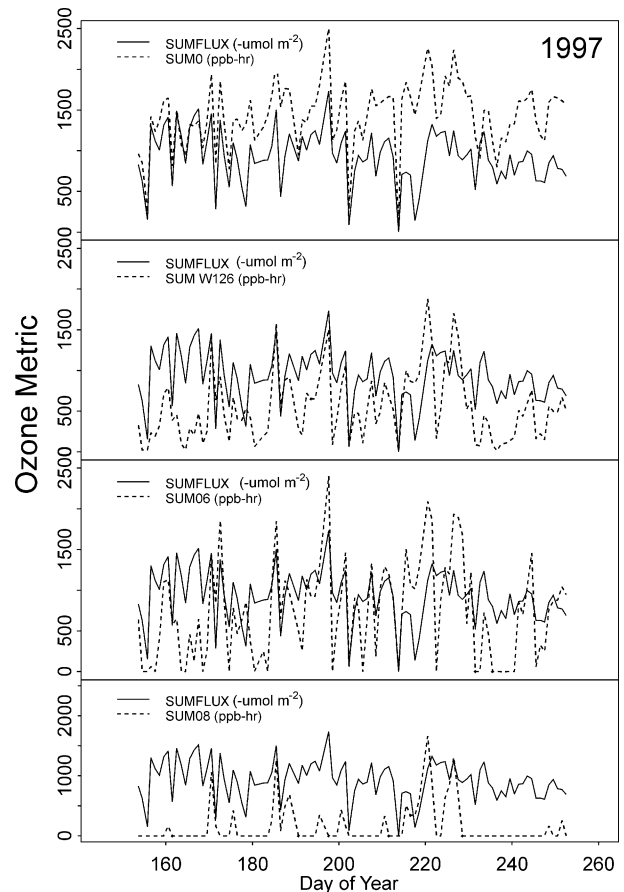


Fig. 6. A comparison of ozone metrics and ozone flux measured over the 1997 growing season.

Ozone concentration based metrics are widely used because ozone concentration is easy to measure and because for some applications, for example irrigated crops, these metrics generally characterize ozone flux well. The need for flux-based metrics, however, is not limited to ecosystems which experience predictable drought, like those in the western United States and the Mediterranean regions of Europe. Variations in water availability from year to year demonstrate the need for flux-based metrics in most non-irrigated systems (Baldocchi, 1997). Furthermore, flux-based metrics are important from a regulatory standpoint as well as a biological perspective. In droughted systems, concentration-based metrics will overestimate ozone uptake, or misrepresent the timing of plant sensitivity to ozone. The use of flux-based metrics might allow for greater efficiency of regulations. Measuring ozone flux directly is expensive and time-consuming, and flux could never be monitored at the spatial scale that concentration is routinely monitored. However, there are methods that could be used to incorporate weighting for physiological activity into concentration-based metrics. In a Mediterranean climate, such as the one described in this paper, weighting concentration metrics by soil moisture would improve the estimate of ozone uptake. A more

accurate means of calculating ozone flux would be to model stomatal conductance and to then estimate fluxes from ozone concentrations and modeled conductances. By employing these more accurate metrics of ozone uptake, our understanding of the relationship between ozone stress and forest response could be significantly improved.

## 5. Conclusion

The metrics that are currently being employed throughout the United States to estimate ozone exposure for forest trees are inaccurate in regions where peak physiological activity does not correspond to peak ozone concentrations. Of the metrics which are commonly used, SUM0 best corresponds to ozone uptake by a ponderosa pine system in the Mediterranean climate of California, however the relationship is only strong early in the growing season when the stomata are unconstrained by drought stress. Over the entire growing season, the time period when these metrics are usually employed, the estimate of ozone uptake by SUM0 may be off by as much as 60%. A better exposure metric would be one that incorporates physiological activity using weighting techniques. Short of directly measuring fluxes, modeled values of stomatal conductance could be used along with ozone concentration data to estimate ozone fluxes.

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